



Review

A review on the sealing structures of membrane electrode assembly of proton exchange membrane fuel cells

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HIGHLIGHTS

- An overview of the sealing structures of MEA was provided.
- The MEA sealing structures were divided into different types.
- The characteristics of each MEA sealing structure type were summarized.

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ABSTRACT

A membrane electrode assembly (MEA) is one of the core components of proton exchange membrane fuel cells (PEMFCs). Its structure is sandwich-like, comprising a proton exchange membrane (PEM), electro-catalyst layers and gas diffusion layers (GDLs). The sealing structure and performance of a MEA are the key factors that influence the performance and the security of fuel cells or fuel cell stacks. In this paper, the sealing structures of MEAs of PEMFCs are reviewed and classified as follows for the first time: (1) PEM direct sealing structure; (2) PEM-wrapped frame sealing structure; (3) MEA-wrapped frame sealing structure; (4) Rigid protective frame sealing structure. The compact MEA-wrapped frame sealing structure has a clearly greater advantage over the other structures for its more convenient stack assembly and lower cost. Another promising sealing structure is the rigid protective frame sealing structure. We hope this review can provide references for the sealing arrangements of PEMFCs available to designers.

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1. Introduction

Due to the energy crisis and environmental deterioration, many countries in the world have intensified their research on new energy sources, with fuel cells being regarded as one of the most promising new energy devices [1]. Fuel cells vary according to the types of electrolyte they employ: proton exchange membrane fuel cells (PEMFCs), alkaline fuel cells, direct methanol fuel cells, solid oxide fuel cells, molten carbonate fuel cells and phosphoric acid fuel cells [2,3]. PEMFCs have been the focus of researchers due to their few moving parts, low operating temperature, high efficiency and environmentally friendly adaptability.

Fig. 1(a) shows the typical structure of a single PEMFC, including the anode and the cathode flow field plates, gas diffusion layers (GDLs), catalyst layers (CLs) and proton exchange membrane (PEM). The fuel cell stack is composed of more than a single cell, as shown in Fig. 1(b). In each cell, the anode CL, GDL, the cathode CL, GDL, and

the PEM are assembled together to form the membrane electrode assembly (MEA), as shown in Fig. 2.

In a MEA, the anode and cathode CL can be coated on one side of each corresponding GDL or on both sides of the membrane. As the key component in fuel cells where the electrochemical reaction takes place and electricity is produced, its performance plays a key role in determining the output and security performance of fuel cells [4]. The performance of a MEA in a fuel cell is decided by the following factors: (1) the characteristics of the key materials, such as the PEM, catalyst and GDLs, etc.; (2) the MEA structure and how it is manufactured; (3) the structure of the flow field plates and fuel cell stack; (4) operating conditions. People have paid much attention to the 1st, 3rd and 4th points, as evidenced by numerous reports in the literature. The MEA should include not only a sandwich-type structure but also sealing features, which prevent the anode fuel and the cathode oxidiser from mixing and stop the gases from leaking to the outside of the fuel cell (stack). In addition, the MEA structures and their size, together with the sealing structures and their size, should align well with the bipolar plates to acquire a suitable clamping force to maintain a favourable contact between the parts, which minimises the electricity resistance,

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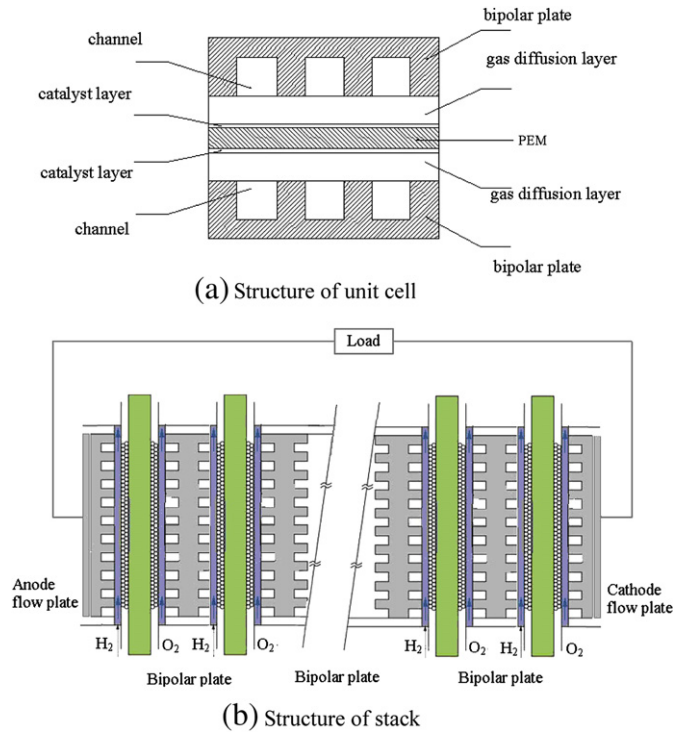


Fig. 1. Schematic diagram of fuel cell.

and to avoid the GDLs from being over compressed, which increases the gases' diffusion resistance when operating under large current density. Fig. 3 shows the contact relationship among the components in a fuel cell.

After a fuel cell stack assembly is completed, the MEA structures and their size, the sealings and bipolar plates should satisfy the following relationship [5]:

$$2d(1 - f_r) - 2C + b_{M_2} = b_{M_1}(1 - f_M) \quad (1)$$

where f_r is the compression value for sealing materials; after this value is reached, effective sealing can be realised. C is the depth of the seal groove in the bipolar plates, d is the diameter of the sealing element, b_{M_1} is the thickness of the MEA corresponding to the sealing parts, b_{M_2} is the thickness of the MEA corresponding to the electrode parts, and f_M is the compression ratio of the MEA under which the minimal contact resistance between the bipolar plates and MEA can be acquired. In Fig. 3, there is no sealing structure preventing the crossover of the anode fuel and the cathode oxidant.

The sealing performance of a MEA is one of the key factors that influence not only the fuel cell assembly efficiency but also the electrical output performance and durability, etc. Many studies have focused on the structures of MEA sealing; however, only a few related reports can be found in the publicly available literature. Lin et al. [6] analysed the chemical degradation of five different sealing

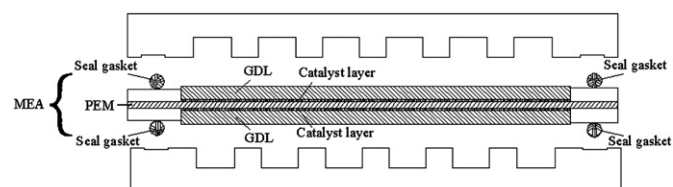


Fig. 2. Structure of membrane electrode assembly.

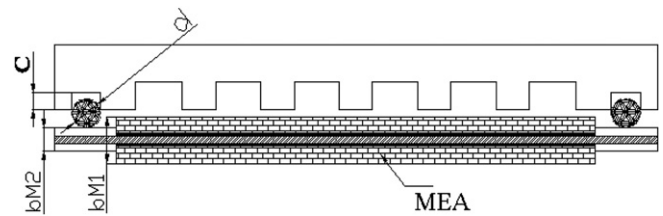


Fig. 3. The contact relationship among the components in a fuel cell. Reprinted from Ref. [5] with permission.

gasket materials under an environment simulating that typical of fuel cell operation. Schulze et al. [7] investigated the degradation of silicone sealing for PEFC test cells during fuel cell operation using XPS and SEM/DEX. The results showed that the decomposition products of the silicone seals had a high mobility, which may contribute to the poisoning of the catalysts. Share et al. [8] investigated the effect of thermal sealing process parameters including sealing temperature, percent compression, seal time and manufacturer-specified post-processing after sealing on fuel cell performance. The results revealed the insignificance of sealing temperature on the initial performance of the MEA and the statistically significant input factors for the thermal sealing process, which were essential for the rapid and high-quality manufacturing of membrane electrode assemblies for high-temperature fuel cells. Bogachev et al. [9] studied the stress distribution and plastic deformation of a MEA during the assembly processes and noted that there was a stress concentration at the junction of the MEA and the sealing gasket. Chien et al. [10] investigated the amount of compression experienced by a PEMFC by measuring it in-situ, i.e., immediately after the assembly and during the normal operation of the fuel cell. The results indicated that the sealing force was not significantly affected during the period between assembly and operation, excluding a stress relaxation effect, and the change in all dimensions due to thermal expansion was negligible. Tan et al. [11] established the relationship between the clamping torques and the level of compression experienced by gaskets in a fuel cell stack by finite element analysis and experiments. Pozio et al. [12] proposed a method using injection moulding to generate the sealing rings of a designed structure at the edge of a MEA and argued that injection moulding could not only reduce the cost but also improve the performance of the seals. Dillard [13] reviewed fuel cell sealings in terms of materials, design and durability.

Due to technical secrets and other factors, more public reports are of patents involving a great variety of sealing structures and materials. This paper reviews the relative literature and tries to classify MEA sealing structures according to their characteristics, mainly based on patents. As in the patents people always try to protect the invention rights meanwhile to keep the core data as secrets, the quantitative information is seldom given, which is a regret.

2. Types of MEA sealing structures of PEMFCs

Researchers have performed many studies on the sealing structures of MEAs. Fig. 4 shows an incomplete summary of related patents and articles about MEA sealing structures published over the past ten years throughout the world, most of which are patents. As shown in Fig. 4, a large number of studies were published each year following 2002. Since then, no paper regarding the classification and review of the MEA sealing structures of PEMFCs has been reported.

To summarise, the sealing structures of MEAs conveniently from the enormous amount of patents that have been published, this

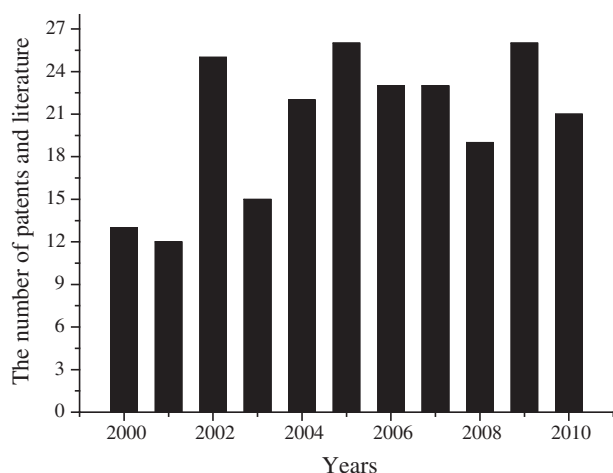


Fig. 4. Published studies on the sealing structures of MEAs.

paper classifies MEA sealing structures as follows: (1) PEM direct sealing structure; (2) PEM-wrapped frame sealing structure; (3) MEA-wrapped frame sealing structure; (4) Rigid protective frame sealing structure. These structure types can be used both for graphite-based bipolar plates, and for metallic bipolar plates.

(1) PEM direct sealing structure. The size of PEMs adopting this structure is larger than that of GDLs; therefore, the PEM is extended beyond the active region and a sealing edge is formed around the cell. Sealing materials such as rubber gaskets [14], PTFE films [15], polyvinylidene fluoride [16], thermoplastic materials [17,18], etc. are placed on both sides of the PEM sealing edge separately. When a fuel cell is assembled, they will be pressed together to form a sealing structure under the clamping force.

(2) PEM-wrapped frame sealing structure. Similar to the structure described above, the size of PEMs adopting this structure is larger than that of GDLs; the PEM is extended beyond the active region and a sealing edge is formed around the cell. The sealing edge of the PEM can be wrapped by adhesive sealing materials, forming the main body of the sealing elements. More typically, the sealing edge of the PEM is placed in a die, into which the moulding sealing materials will be injected. After completing moulding, the sealing materials will be shaped and form the main body of the sealing structure. This kind of sealing structure becomes an indivisible part of the MEA, i.e., an integrated MEA with a sealing structure. Because of these features, it can prevent gases both from leaking to the outside of the fuel cell and from crossing over to other areas of the cell.

The various materials used for injection moulding include thermo-plastic/thermosetting polymers, vulcanised rubber and cold curing resin as well as polymerisable oligomer polypropylene polydienes such as polybutadiene and polyisoprene.

(3) MEA-wrapped frame sealing structure. Here, MEA refers to the membrane electrode, including the membrane, the catalyst layers and the GDLs, without other sealing materials. The non-active region around the active part of the MEA forms the sealing edge. Similar to the second type of structure described above, this sealing edge can be wholly or partly wrapped by adhesive sealing materials, or more commonly wrapped by injection moulding sealing materials, after which the shaping of the integrated MEA with the sealing structure is completed.

The materials used for injection moulding include thermo-plastics, solidifying sealants and adhesives, such as liquid crystal polymers (LCPs), polyphenylene sulphide resin, polysulphone (PSF) [19], silicone [20,21], polyurethane, elastomeric materials, vinylidene fluoride [22], polyurethane [23], epoxy resin, photoinitiators, etc.

(4) Rigid protective frame sealing structure. In this sealing structure, the frame around the edge of the MEA is composed of rigid materials, such as polyethylene naphthalate (PEN) [24], polyethylene terephthalate (PET) [25], Teflon [26], polytetrafluoroethylene (PTFE) [27], polyvinylidene fluoride (PVDF), ethylenevinylalcohol (EVOH), biaxially oriented polypropylene (BOPP), polyether sulphone (PES), fluorinated ethylene propylene (PEP), polyimide (PI) [28] etc., which form the main body of the sealing structure after being hot-pressed with other melting and binding thermo-plastic materials and sealing materials.

The structure and the size of the rigid frame determine the compression rate of the MEA after the fuel cell stack is assembled. After compression, the structure can perform a sealing function, maintaining a minimal contact resistance between the MEA and bipolar plates while making sure the MEA is not over compressed to cause mass transfer problems during operation or even become damaged.

Fig. 5 shows what fraction of all MEAs considered feature each kind of sealing structure. No one MEA sealing structure holds an absolute advantage over the others, but we can trace the developing trend according to type and prevalence in the literature. This is discussed in detail in the following section.

3. MEA sealing structures of PEMFC

3.1. PEM direct sealing structure

PEMs are larger than GDLs in this sealing structure; the sealing materials are compressed directly on the non-active marginal regions of the membrane [29–46]. The fuel cell sealing function is achieved by pressing the sealing rings tightly, as shown in Fig. 6.

Fig. 6a shows one MEA sealing structure proposed by Yang et al. [29]. They used sealing gaskets that were not destructive to the PEM and arranged them along the outer edges of the PEM of the

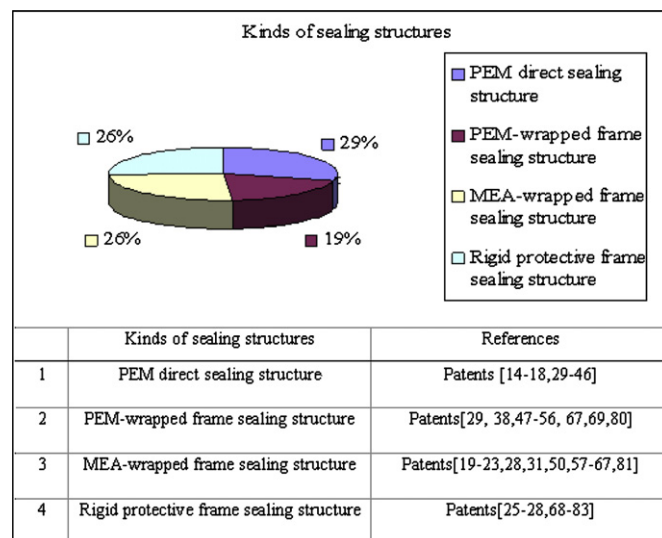


Fig. 5. Four different kinds of MEA sealing structures based on the patents.

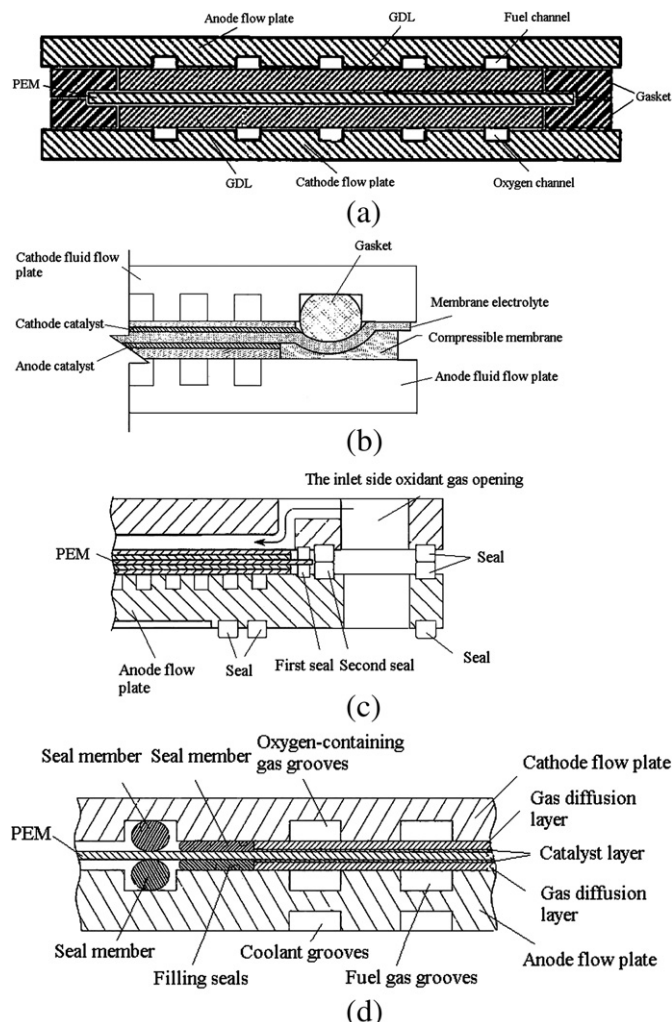


Fig. 6. PEM direct sealing structure (a–d). Reprinted from Ref. [29–32] with permission.

cathode and anode sides of the PEM. Bipolar plates with flow channels were adopted to compress the seal gaskets tightly; therefore, the gases in the anode and cathode were separated from each other and also from the outside.

Matlock et al. [30] set sealing grooves on the cathode plate, into which the sealing rings were placed, while on the anode side, they used a kind of compressible component. The contact surface shape between these components and the PEM made allowed the PEM to be positioned rigidly, in addition to being sealed, as shown in Fig. 6b.

Inoue et al. [31] used a double sealing pattern in a fuel cell assembly, as shown in Fig. 6c. The first set was on both sides of the PEM, which was compressed on the membrane directly and formed the first sealing to prevent the gases from crossing over. The second set was between the MEA and bipolar plates, forming the second sealing to prevent the gases from leaking to the outside.

Yoshida et al. [32] used a double sealing pattern similar to the one described above but with differences regarding certain details as shown in Fig. 6d. In the first sealing, both sides of the PEM were coated with sealing binder, while the other gaskets were arranged in the bipolar plate grooves to clamp the PEM to create the second sealing. The two sealing materials were different, but both acted on the PEM directly and had the same sealing functions.

Hence, the fundamental features of the direct PEM sealing structure are as follows: (1) the MEA and sealing components are

fabricated before being integrated into fuel cells. As a whole, this sealing structure is simple and is suitable for frequent reassembly. (2) The membrane extends to the non-active region, which forms the sealing edge. The demand for preventing gases from leaking internally mainly depends on membrane permeability, and the seal gaskets play key roles in restraining the gas from leaking to the outside. (3) The sealing materials directly contact the membrane; therefore, they should not effect any damage on the membrane. (4) The sealing gaskets in the anode and cathode clamping the membrane may lead to membrane breakdown due to the shear and tear generated at the edges of the seal gaskets, thus causing the seal to fail.

3.2. PEM-wrapped frame sealing structure

PEMs in this structure are larger than GDLs; the PEM is extended beyond the active region and a sealing edge is formed around the cell [47–56].

One version of this sealing structure is designed such that the sealing edge of the PEM can be wrapped by adhesive sealing materials, forming the main sealing elements. Schmid [52] introduced a structure in which an adhesive sealing material was used to wrap the edge of the PEM and bond the two bipolar plates adjacent to the MEA; together, all of the components constructed one unit cell module, as shown in Fig. 7a. In this sealing structure the internal leaking of gas from anode to cathode and vice versa was prevented by the membrane as well as the adhesive in the sealing gaskets, the latter of which restrained the gases from leaking to the outside. The integrated unit cell module makes it easy to assemble fuel cell stacks, but the adhesiveness of the sealing material makes it difficult to disassemble a stack and then reassemble it.

Generally, the PEM-wrapped frame sealing structure is a PEM plastic frame. The PEM is larger than the GDL. The sealing edge of the PEM is held by a die, and the plastic material is then injected into the die, after which the injected material becomes the main sealing elements.

Adachi [53] first proposed this structure in 1999, as shown in Fig. 7b. The sealing materials plasticised at the sealing edge of the PEM and were integrated into the MEA as an elastic frame, which performed the sealing function after being compressed when the fuel cell was assembled.

Similar to the sealing structure frame shown in Fig. 7b, another structure introduced by Suenaga T. et al. [54,55] (Fig. 7c) features a frame with a different cross-sectional shape. Fig. 7c also shows the die structure. In this sealing structure, the edge of the PEM was not easy to fasten; thus, the PEM might be deformed or self-crimped, leading to a weakened sealing effect. To improve this condition, Takada K. [56] drilled a small hole at the edge of the PEM and fixed the PEM onto the projections of the die; thus, the sealing effect was better than that of the previous structure, as shown in Fig. 7d.

To summarise the main features of the PEM-wrapped frame sealing structure are as follows. (1) After the sealing structure is injection moulded, the active area of the MEA and the sealing structure are integrated together into a single component, i.e., the integrated MEA, which is not only beneficial for stack assemblies but also suitable for mass production. (2) The membrane extends to the non-active sealing area, and the internal leaking of gas from anode to cathode, or the opposite, is prevented by the membrane and the sealing gaskets shaped by injection moulding, latter of which restrain the gases from leaking to the outside. (3) The materials used for injection moulding include a variety of thermoplastic/thermosetting polymers, vulcanised rubber and cold curing resin as well as polymerisable oligomer polypropylene polydienes such as polybutadiene and polyisoprene. (4) The sealing elements shaped by injection moulding must satisfy not only the sealing

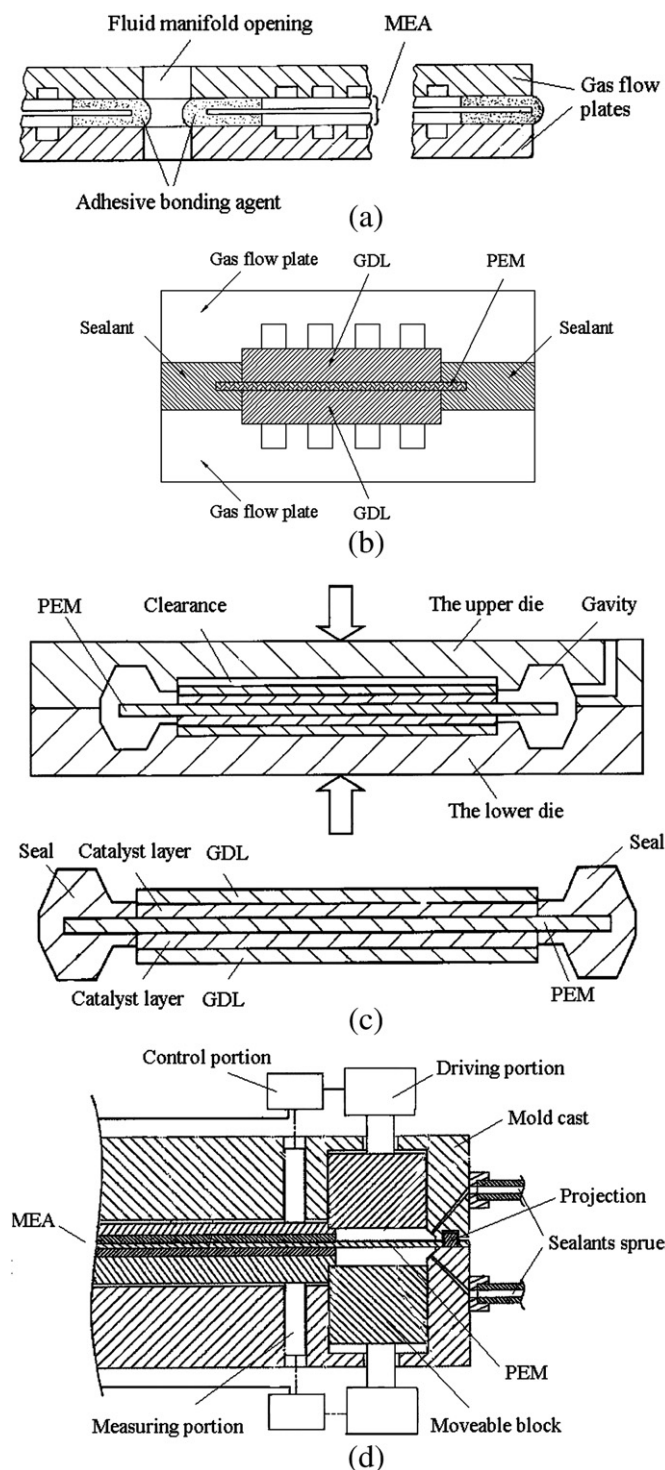


Fig. 7. PEM-wrapped frame sealing structure (a–d). Reprinted from Ref. [52–56] with permission.

function requirements but also optimise the contact pressure to acquire a minimum contact resistance while preventing the GDLs from being over compressed. The working temperature and pressure in the injection moulding process should not be so high as to damage the membrane. Therefore, there are rigorous requirements for the die structure and size, the characteristics of the sealing materials and the process parameters. (5) The sealing frame shaped by injection moulding wraps only the edge of the PEM; the joints

between the frame and the GDLs may be a potential problem. (6) Some adhesive materials can be used to wrap the PEM directly and bond the bipolar plates together to construct a single integrated fuel cell, which makes it easy to assemble a stack but difficult to dismantle and reassemble it many times.

3.3. MEA-wrapped frame sealing structure

The main features of this structure are that the sealing frames wrap the sealing regions of the MEA, which may include the PEM, the CLs and GDLs on both sides of the PEM; in some cases, the frame may wrap only the PEM and part of the material layers on both sides or one side of the PEM [19,21,22,57–67].

Similar to the one sealing structure described in Section 3.2, the non-active areas of the MEA in this sealing structure can be entirely or partly wrapped by adhesive sealing materials, which compose the main sealing components. Wozniczka et al. [57] bonded the sealing edge of MEA and the sealing area of the flow field plates using an adhesive. In addition, manifold sealing members were set around the MEA, which formed the second sealing elements, as shown in Fig. 8a.

More commonly, the non-active portions of the MEA are positioned in the die; then, the injection moulding materials are

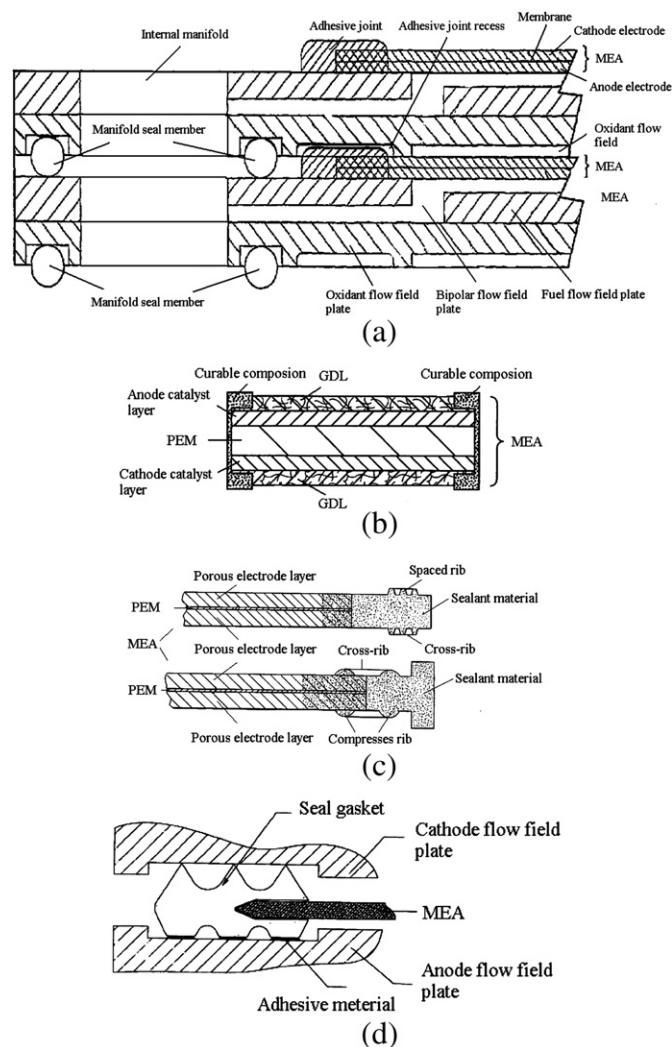


Fig. 8. MEA-wrapped frame sealing structure (a–d). Reprinted from Ref. [57–60] with permission.

injected into it, and the principal parts of sealing components are formed.

Jacobine et al. [58] injected light-cured sealants into transparent dies to wrap the sealing edge of MEAs to construct the sealing frames, as shown in Fig. 8b. This is also a typical integrated MEA structure with sealing features. It should be noted that the sealing materials have to permeate the porous GDLs and/or the CLs and can also form the soft sealing frames, unlike in the PEM wrapped frame sealing structure. Therefore, the selection of the sealing material and the process parameters is also very strict.

Barton et al. [59] constructed a sealing frame around a MEA. The sealing material permeated porous media such as the GDL during injection moulding and formed the integrated MEA structure. To improve the sealing function, the cross sections of the frames were designed as various structures, as shown in Fig. 8c.

The methods described above are implemented by injecting sealing materials into dies; thus, highly precise dies are required. In addition, the high temperature and pressure required for injection moulding may damage the MEAs.

To overcome some of these shortcomings, Hu et al. [60] prefabricated a kind of sealing gasket using a die, as shown in Fig. 8d. The two sides of the gasket had saw-toothed features; it also had a mouth opening into which the sealing edges of the MEA were placed. After a fuel cell is assembled using this structure, the compressing force supplied by the bipolar plates would compel the gasket to hold the MEA and realise the sealing functions between the two sides of the MEA and fuel cell units.

To summarise, the main features of the MEA-wrapped frame sealing structure are as follows. (1) After the sealing structure shaped using adhesive wrap, light-cured materials, or injection-moulded materials, the active area of the MEA and the sealing structure are integrated together into a single component, i.e., integrated MEA, which is not only beneficial for stack assemblies but also suitable for mass production. (2) The materials used for injection moulding include thermoplastics, solidifying sealants and adhesives such as liquid crystal polymers (LCPs), polyphenylene sulphide resin, polysulphone (PSF), polyether-ether-ketone (PEEK), polybutylene terephthalate (PBT), polyvinylidene fluoride (PVDF), polyolefin, rubber, polyurethane, epoxy resin, photoinitiator, etc. (3) Under the effects of pressure and temperature, the sealing materials may melt and penetrate into porous media, such as the non-active area of GDLs and/or CLs, which makes the joints tighter and more even between the sealing frame and the active part of the MEA. (4) The adhesive material can be used to wrap the edge of the MEA directly to form the sealing frames; this is inconvenient for the disassembly and reassembly of fuel cells. (5) Sealing frames can be prefabricated and assembled together with MEAs.

3.4. Rigid protective frame sealing structure

The basic idea of the rigid protective frame sealing structure is that there is a frame around the edge of the MEA [68–83], which is made of rigid materials, such as PEN, PTFE, Teflon, etc. it will form the main body of the sealing structure after being hot-pressed with other melting and binding thermo-plastic materials and sealing materials. The structure and the size of the rigid frame determine the compression rate of the MEA after the fuel cell stack is assembled; after being compressed, it can play a sealing function and maintain a minimal contact resistance between the MEA and bipolar plates while making sure the MEA is not over compressed to cause mass transfer problems or even become damaged during operation.

Masaaki et al. [74] introduced a step-type sealing structure in which the membrane was fixed on the step using a binder to protect the fuel and the oxidant from crossing over, as shown in Fig. 9a.

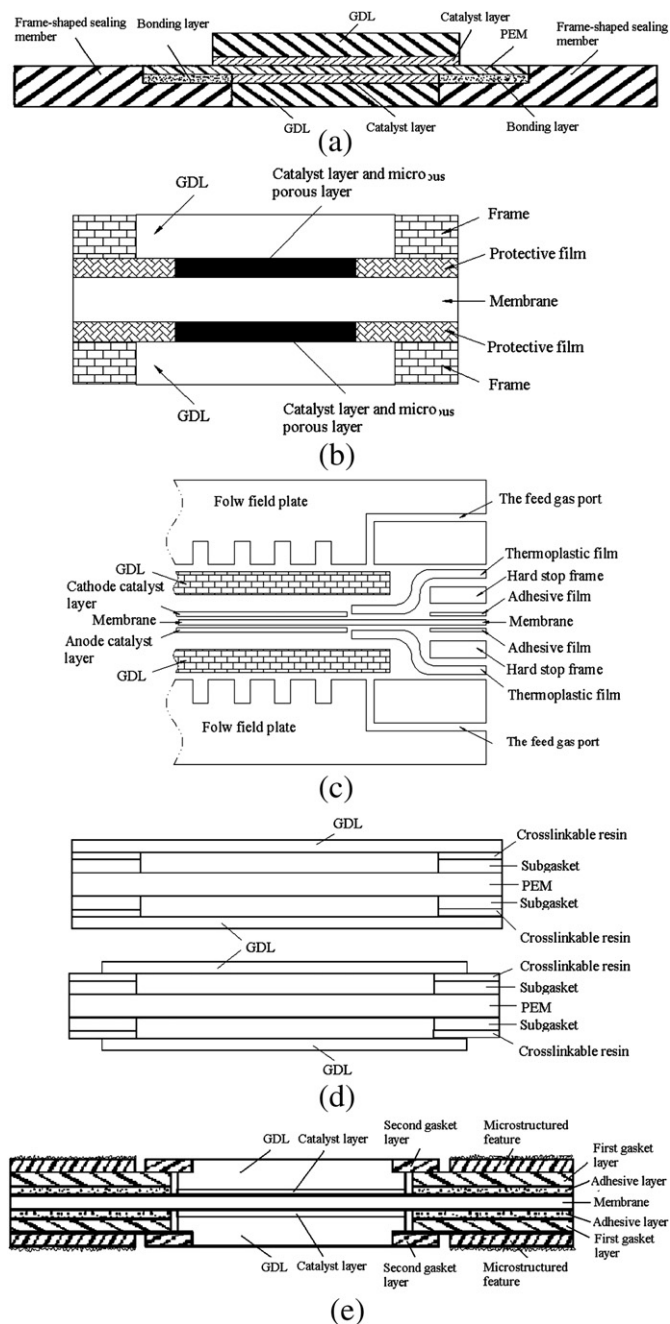


Fig. 9. Rigid protective frame sealing structure (a–e). Reprinted from Ref. [74–78] with permission.

The frame could be composed of rigid resin materials, which were observed to support the excessive pressure when the optimal compression rate of MEA was reached; therefore, the MEA was protected from being over compressed when the fuel cell was assembled. Other rubber sealing gaskets were needed on the surfaces of the rigid frame to prevent the gases from leaking to the outside.

Xu et al. [75] placed a kind of protective film, such as a PTFE film, on each side of the PEM in the non-active region; another hard frame was placed on the outside of this film on each side, as shown in Fig. 9b. The thickness of the film was about the same as that of the porous carbon layer. Both the film and the frame were almost incompressible; therefore, they supported the compression force

derived from the bipolar plates when the fuel cell stack was assembled. This structure could prevent the active area of the MEA from being over compressed or even damaged. Other rubber sealing gaskets were needed on the surfaces of the hard frame to avoid the gases from leaking to the outside from the gap between the frame and the bipolar plate.

Mao et al. [76] placed so-called hard stop frames on both the anode and cathode sides of the sealing edge of a PEM. In each side, a layer of thermoplastic material was used to glue the bipolar plate and one side of the hard stop frame; an adhesive film was used to glue the PEM and the other side of the hard stop frame, as shown in Fig. 9c. The hard stop frame sustained the main compression force from the bipolar plates when the fuel cell stack was assembled, which prevented the MEA from being over compressed or even damaged. As the thermoplastic film and the adhesive film connected the fuel cell parts and the sealing part together, it was difficult to disassemble and reassemble the fuel cell stack repeatedly.

As shown in Fig. 9d, Yandrasits et al. [77] placed a protective subgasket around the membrane on each side; the subgasket was composed of a certain hard material. A crosslinkable resin was placed between the GDL and the subgasket; the resin could be light-cured or thermal-cured. After processing, the MEA and the sealing parts were connected to form a single unit.

Debe et al. [78] made many improvements in the structural details, as shown in Fig. 9e. An adhesive layer was placed between the non-active membrane edge and the first gasket. Another layer with a micro concavo-convex structure was placed on the first gasket to prevent gas leakage during fuel cell assembly. A bonding material was used to fill the gap between the GDL and the microscopic structure, which integrated the first gasket layer and GDL after being heated (pressurised), thus constituting the second border layer. The integration of the membrane electrode and frame enhanced the stability of the overall structure.

In conclusion, the main features of the rigid protective frame sealing structure are as follows: (1) One or more hard layers were set on both sides of the non-active membrane edge, which sustained the main compression force from the bipolar plates during fuel cell assembly, therefore preventing the active area of the MEA from being over compressed or being damaged. (2) The hard sealing frames, adhesive materials, and membrane are usually agglutinated together to avoid fuel and oxide crossover; other rubber gaskets are needed to prevent the gases from leaking to the outside. (3) The overall structures are complex, and the structure and the size of each component should be exactly matched with one another to satisfy the requirements of both sealing and fine contact.

4. Conclusions

The sealing structure and performance of MEAs are two of the key factors affecting the electricity output performance and security of fuel cells. None of the MEA sealing structures has an absolute advantage over the others. The development trend observed in recent years indicates that the reasonable design of the MEA sealing structure should be suitable for mass production, convenient for fuel cell stack assembly and low cost, that is, an integrated MEA with sealing structures. Therefore, the compact MEA-wrapped frame sealing structure created by injection moulding has a clearly greater advantage over the other structures. The one-time-shaped sealing structure has strict requirements regarding die structure and size, the materials used for moulding, and the process parameters.

Another promising sealing structure is the rigid protective frame sealing structure. Its main characteristic is that one or more hard

layers are set on both sides of the non-active membrane edge, which sustain the main compression force derived from the bipolar plates during fuel cell assembly and prevent the active area of the MEA from being over compressed or damaged. However, this sealing structure is complex and it consists of many components, including the rigid frames and adhesive layers, which require prefabrication before MEA processing. Thus, the material characteristics and the structures and the sizes of all components should be chosen and designed carefully.

It can be concluded that increasingly more reasonable and suitable MEA sealing structures will be created as a result of ongoing research, which will promote the development of fuel cell technology and commercialisation.

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